

## On thermoluminescence glow curves with second order kinetics

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*Received 20 February 1996, accepted 2 May 1996*

**Abstract** : We have made a comparative study of the conventional second order model of thermoluminescence (TL) with that of Prakash [1]. We conclude that the model of Prakash is unable to explain characteristics features of second order TL.

**Keywords** : Thermoluminescence, order of kinetics, symmetry factor.

**PACS No.** : 78.60.Kn

Recently Prakash [1] has proposed a theory of second order thermoluminescence (TL) glow curves. According to the existing theory [2,3] for second order TL peaks, the TL peak temperature  $T_m$  decreases with increasing  $n_0$ , the initial number of trapped charge carriers. But citing some earlier works [4,5] Prakash claims the aforesaid dependence of  $T_m$  on  $n_0$  to be an unexpected behaviour. The shift of  $T_m$  with  $n_0$  is manifested in the shift of  $T_m$  with irradiation dose [6]. Pokorny and Ibarra [7] observed the shift on  $T_m$  with irradiation dose for TL of several  $\text{Al}_2\text{O}_3$  samples doped with different impurities mainly Cr and Ni. More recently, Gartia *et al* [8] also observed the shift of  $T_m$  with dose. So it is seen that the starting point of the second order TL model of Prakash [1] *i.e.* the peak temperature  $T_m$  does not show dependence on  $n_0$ , contradicts the experimental findings.

In order to explain the 'independence' of  $T_m$  and  $n_0$  but at the same time the dependence of the peak intensity  $I_m$  on  $n_0$ , Prakash [1] proposed a new model of second order TL by modifying the existing Adirovitch [9] equations with the introduction of an *ad hoc* parameter  $x = (n/n_0)^{b-1}$  in the charge neutrality condition. Here  $b$  is the order of kinetics and  $n$  is the density of electrons in the trapping levels. According to Prakash, the charge neutrality condition is given by

$$m = xn + n_c, \quad (1)$$

where  $m$  represents the density of holes in the recombination centres and  $n_c$  the density of electrons in the conduction band. However, no theoretical justification for arriving at eq. (1) has been given. The equation for the glow intensity arrived at by Prakash for a TL glow curve with order of kinetics  $b$ , can be written as [1]

$$I(T) = b n_0 s \exp(-E/kT) \exp\left[-(bs/\beta) \int_{T_0}^T \exp(-E/kT') dT'\right], \quad (2)$$

where  $s$  is the frequency factor,  $E$  the activation energy and  $\beta$  the linear heating rate.  $T_0$  and  $T$  are respectively the starting temperature and temperature at time  $t$  in  $K$ . The maximum condition is given by

$$E/kT_m^2 = (bs/\beta) \exp(-E/kT_m). \quad (3)$$

Eliminating  $bs/\beta$  from eqs. (2) and (3) by following Christodoulides [10] one can write

$$I/I_m = \exp[u_m - u + F(u, u_m)] \quad (4)$$

with  $F(u, u_m) = u_m^2 [E_2(u_m)/u_m - E_2(u)/u] \exp(u_m)$ ,

where  $E_2(u)$  is the second exponential integral [11] and  $u = E/kT$ ,  $k$  being the Boltzmann constant.

It is evident from eq. (4) that the fractional intensity  $x = I/I_m$  in Prakash model for any order of kinetics is identical with that of first order kinetics ( $b = 1$ ) model of Randal and Wilkins [12]. So following the work of Gartia *et al* [13], the symmetry factor [14]  $\mu_g(1/2)$  at the half intensity points in Prakash model will lie between 0.446 and 0.408 as  $u_m$  changes from 10 to 100. This range of  $u_m$  covers almost all the observed cases of TL [10]. So the model of Prakash can not explain the values of  $\mu_g(1/2)$  namely 0.475 and 0.520 as observed by Prokic [4] for 478K and 520K peaks of natural barite although he cites the work of Prokic [4] in support of his model.

In Table 1 we compute  $\mu_g(1/2)$  of some numerically computed second order TL peaks corresponding to Figure 1 of Chen *et al* [3]. We note that Figure 1 of Prakash [1] is almost identical with that of Chen *et al* [3]. As a check of our computer code we have reproduced the peak temperatures  $T_m$  following the technique outlined by Singh *et al* [15]. It is clear from Table 1 that the values of  $T_m$  are in excellent agreement with those of Chen *et al* [3]. We also see from Table 1 that the calculated values of  $\mu_g(1/2)$  are very close to

the conventional second order value namely 0.52 [14]. Thus, the model of Chen *et al* [3] unlike that of Prakash [1], can account for the observed value 0.52 of the symmetry factor for the second order 520 K TL peak of natural barite [4]. Similar computation along the lines suggested by Singh [16] by using the general order extension [17] of the model of Chen *et al* [3] can explain the occurrence of the value 0.475 for the 478 K TL peak of barite having order of kinetics 1.5.

**Table 1.** The values of peak temperature ( $T_m$ ) and symmetry factor  $\mu_g(1/2)$  for a set of numerically computed second order TL glow curves (Chen *et al* [3]) with different initial concentrations ( $n_0$ ) of trapped electrons for  $E = 1.1$  eV,  $\beta = 1$  K/s and  $s' = 10^{-6}$  m<sup>3</sup> sec<sup>-1</sup>  $a(b)$  stands for  $a \times 10^b$ .

| $n_0(\text{m}^{-3})$ | $T_m(\text{K})$       |         | $\mu_g(1/2)$ |
|----------------------|-----------------------|---------|--------------|
|                      | Chen <i>et al</i> [3] | Present |              |
| 2.0 (16)             | 479.8                 | 478.6   | 0.520        |
| 4.0 (16)             | 466.7                 | 467.3   | 0.520        |
| 8.0 (16)             | 455.9                 | 456.5   | 0.519        |
| 1.6 (17)             | 446.4                 | 446.2   | 0.519        |
| 3.2 (17)             | 436.3                 | 436.4   | 0.519        |
| 6.4 (17)             | 428.6                 | 426.9   | 0.518        |

We note another misconception in the work of Prakash [1] in connection with the work of Sinha and Mukherjee [5] on TL of  $\text{CaF}_2 : \text{Pr}$  single crystals. According to Prakash the fact that the location of 530 K TL peak in  $\text{CaF}_2 : \text{Pr}$  is independent of the initial concentration of Pr in  $\text{CaF}_2$  as observed by Sinha and Mukherjee [5] indicates that the peak temperature  $T_m$  is independent of  $n_0$ . But this contention is not correct. Sinha and Mukherjee [5] state that the 530 K TL peak in  $\text{CaF}_2 : \text{Pr}$  shifts towards the higher temperature with the increase of thermal cleaning temperature of adjacent peaks. This fact is an evidence of the shifting of  $T_m$  with  $n_0$  [6]. Again the comment of Prakash that the second order model of Chen *et al* [3] gives different values of  $E$  and  $s$  for various samples of the same system is not appropriate. All the parameters  $E$ ,  $s$ ,  $b$  and  $n_0$  of a general order TL peak can be determined by using a rigorous computer code of curve fitting as done by Singh [16] and Singh and Gartia [18] for the TL of loess [19].

We conclude that the second order TL equation suggested by Prakash [1] is in fact equivalent to the first order TL equation of Randal and Wilkins [12]. As a result, it fails to explain the experimental findings [7,8] on the shift of TL peak temperature ( $T_m$ ) with the irradiation dose and the symmetry factor in the case of second and other non-first order glow curves [4].

### Acknowledgments

Thanks are due to Professor R K Gartia for fruitful discussions. One of us (NCD) thanks Department of Science and Technology, Government of India for financial assistance.

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